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MOISTURE TRANSFER IN SOLID FOOD MATERIALS: A REVIEW OF MECHANISMS, MODELS, AND MEASUREMENTS

Jaruk Srikiatden

Department of Food Science and Technology, Cornell University, Geneva, NY, USA

John S. Roberts

Western Regional Research Center, Agricultural Research Service, United States Department of Agriculture, Albany, CA, USA

Drying is one of the most important processes in the food industry, as well as one of the most frequently studied topics in food engineering. Optimizing this process will result in lower production costs and increased product quality. Moisture transfer in heterogeneous materials, such as foods, is a complex process where more than one mechanism may occur. After over eighty years of applying Fick's Second Law diffusion equation to drying of foods, there are still wide variability in reported diffusion coefficients. This article reviews moisture transfer mechanisms, models developed to predict moisture transfer, measurement of effective moisture diffusivity, which is the most common parameter used in predicting moisture transfer, and advanced measurements of moisture profiles to quantify and validate predictive models.

Keywords: *Dehydration, Models, Mechanisms, Diffusivity.*

INTRODUCTION

Dehydration is a significant unit operation in the food industry, with products ranging from sugar, coffee, cornstarch, flour mixes, dried fruits, breakfast food, pasta products, baked goods, and many others. Drying process extends shelf life of perishable foods, such as fruit and vegetables, by removing water to a level at which microbial spoilage and chemical reactions are minimized.[1,2,3] Due to an increasing resistance to the use of chemicals as food preservatives, high quality dried foods with good rehydration properties are a renewed interest in drying operations.[4] In addition to preservation, drying of food products is intended to improve product stability, decrease shipping weights and costs, and minimize packaging requirements.[5,6] A substantial fraction of the energy consumed in process industries is required for drying operations.[7] Therefore, understanding internal moisture transfer mechanisms are important to optimize both the quality of the product and the economics of the process. This review has 3 main sections describing moisture

transfer mechanisms, common drying models used along with their underlying assumptions, and methods used to measure effective moisture diffusivity. In addition, physical characteristics and moisture profile methodologies are described.

PHYSICAL CHARACTERISTICS OF THE MATERIAL

The mechanisms by which heat and mass are transferred in a material depend on the physical structure and chemical composition of the material.[2] A system can be classified according to its water binding characteristics, such as non-hygroscopic or hygroscopic. A material that is non-hygroscopic, such as glass wool or ceramic, does not contain bound water where the partial pressure of the water in the material is equal to the vapor pressure of free water.[8] A material that is hygroscopic contains bound water, and the vapor pressure of the material is less than that of free water. Foods, in general, can be considered hygroscopic. Of course, there are exceptions, as is likely in any classification process. For instance, a hygroscopic material with an initial equilibrium moisture content above atmospheric saturation would have a vapor pressure equal to that of pure water. Therefore, this particular hygroscopic material would behave as a non-hygroscopic material.[9] Non-hygroscopic materials may also have characteristics of hygroscopic materials. Sand, for instance, is commonly considered a non-hygroscopic material; however, when its equilibrium moisture content is very low (less than 0.1 g/g d.s.), sand does have bound water. Since this transition occurs at very low equilibrium moisture contents, sand is generally considered a non-hygroscopic material.[10] The relationship between moisture content and partial pressure as a function of temperature is commonly expressed in sorption isotherms.[9,11] Knowledge of sorption behavior of food is useful in drying processes because it can be used to predict the optimum drying time and final moisture content of the product.[11,12]

The porous structure of dehydrated plant products has an important effect on quality characteristics and the transport properties of these products.[13,14] Porosity is defined as the ratio of the free space occupied in the material to the total volume of the material.[15] What defines a porous and a non-porous material still remains to be determined, though there are a few studies that suggest a non-porous material has porosity less than 0.25 and a porous material would have a porosity greater than 0.4.[15–21] Goedeken and Tong[16] conducted permeability measurements on pregelatinized flour dough as a function of porosity between 0.10–0.60, where porosity was controlled by chemically leavening using sodium bicarbonate. The results showed that permeability values ranged from 1.97×10^{-14} m^2 (0.02 darcies) at 0.10 porosity to 2.27×10^{-11} m^2 (23 darcies) at 0.60 porosity, and the permeability was low (7.9×10^{-13} m^2 (< 0.8 darcies)) in the bread samples having a porosity less than 0.4. The samples having porosity less than 0.4 had less interconnecting pores resulting in an increase in resistance to air flow through the samples, and thus a material with a porosity less than 0.4 could be classified as non-porous. Moreover, Waananen and Okos[17] reported that a material is considered porous when its porosity is above 0.25. Porous food materials would include baked products such as white bread and butter cookies, with porosities of 0.90 and 0.55, respectively.[18] Powdered products would also be considered porous, such as flour, milk powder, and chocolate pudding powder with porosities of 0.69, 0.454–0.61, and 0.5, respectively.[18] The inner-connecting pores in a solid would certainly differ to those in granular materials;[16] therefore, the definition of a porous material would need to be defined separately for solid and granular materials. Examples of non-porous, or dense food materials are potato, carrots, and apples having porosities of 0.02, 0.04, and 0.21, respectively.[15] Increase in bulk porosity of some dried

materials indicates that pores were developed during the drying process, which can significantly affect the moisture transfer properties or the mechanism of transfer. Carrots and potatoes develop almost negligible porosity (<10%), whereas apples develop significant porosity (0.20 to 0.70) during drying.[19,20,21]

As with any classification, there are limitations and exceptions particularly when we place the definitive "non" in front of the description. So as described above, non-hygroscopic materials may have some water binding properties, such as sand, and non-porous foods have some pores, such as potatoes. Analogous to this water binding and structure classification is the widely used thermal and non-thermal processing in the field of food engineering and processing. Of course non-thermal processing does not mean we are processing at absolute zero. The terminology is relative, such as the overall water binding characteristics, fraction of pore space, or processing at temperature below lethal temperatures for target microorganisms and deterioration of enzymes and nutrients are minimized. Perhaps a better terminology would be to use "low," "mid" or "intermediate," and "highly" to classify levels of hygroscopic and porosity instead of "non." Thus, based on research mentioned previously, a low porous structure would have a porosity below 0.25, a intermediate porous structure would have a porosity between 0.25 and 0.4, and a highly porous structure would have a porosity above 0.4. Hygroscopic classifications would be based on the material's sorption isotherm.

Most hygrscopic materials shrink in volume upon drying, which may alter the mechanism by which moisture or vapor migrates through the material. The shrinkage during drying is usually linearly proportional to the change in moisture content.[15,19,20,22] The shrinkage of porous materials during the drying stage is dependent on the internal vapor pressure.[23] The physical properties of a material, such as density and porosity, are also affected by shrinkage during drying,[24] but to what degree depend on the material being dried. Porous foods tend to shrink less than non-porous foods during drying due to the fact that porous foods normally have less initial moisture content than non-porous foods. Bread, for instance, has been shown to only shrink by 10% during drying.[25] In the case of materials with high moisture content, microstructural stresses induced by moisture gradients within the material results in shrinkage.[26]

MECHANISMS OF MOISTURE TRANSFER

Drying can be divided into a constant rate period and one or more falling rate periods.[9,11] Constant-rate drying is defined as the period of drying where moisture removal occurs at the surface by evaporation and the internal moisture transfer is sufficient enough to maintain the saturated surface, thus the rate of evaporation remains constant. The rate of energy input equals the heat lost during evaporation, so the temperature at the surface is also constant and is lower than the surrounding temperature of the air at a wet-bulb temperature. The rate of moisture removal is controlled by the rate of evaporation of moisture from the surface to the drying medium as well as the rate of heat transfer to the evaporating surface. Therefore, drying in the constant-rate period is a surface-based rate governed by external conditions such as temperature difference between the dry air and wet surface, area exposed to the dry air, and external heat and mass transfer coefficients.[8,27]

The transition moisture at which the departure from constant rate drying is first observed is called the critical moisture content, M_c.[11,27] The critical moisture content usually varies with the thickness of the material and the external drying conditions. Saravacos and Charm[28] determined the critical moisture content of some fruits and vegetables and

found that M_c of fruits varied from 5.5 to 7.7 g/g dry solid and M_c of vegetables varied from 3.5 to 5.0 g/g dry solid. These reported critical moisture contents are close to the initial moisture contents, indicating the decreasing importance of the constant rate period in food dehydration. This is confirmed by the absence of constant rate periods during drying of many fruit and vegetable products, such as tapioca, sugar beet root and avocado as reported by Chirife and Cachero,[29] Vaccarezza et al.,[30] and Alzamora and Chirife,[31] respectively. Most drying of biological products occurs during the falling-rate period.[28,30,32]

The falling-rate period is characterized by the surface of the material not being saturated, and the rate of moisture movement from the interior towards the surface is less than the rate of evaporation from the surface, thus drying in the falling rate period is an internally controlled mechanism.[27,33] In the first falling-rate period, the drying rate decreases as the moisture content decreases due to the additional internal resistance for moisture transfer and to the reduction of heat flux into the sample as the surface increases to the heat medium.[9,11] Though some regions of the sample become porous from loss of moisture, a large portion of the sample remains non-porous during the early part of this period. The temperature at the interior of the sample does not increase significantly from the wet-bulb temperature. The second falling-rate period begins when the partial pressure of water throughout the material is below the saturation level. The heat flux from the hot air to the sample is very low because of the small temperature gradient between the hot air and the surface, which is now close to the hot air temperature.[8] In a highly porous material or when significant porosity is developed, mass transfer occurs mainly in the vapor phase and all evaporation occurs from the interior of the material.[11] Furthermore, in order to supply the heat of vaporization to the interior, heat has to be conducted through dry solid and pore regions, both of which have low thermal conductivity. The drying rate in this period is extremely slow; therefore, it is not surprising that the time required to remove the last 10% of moisture is almost equivalent to the time required to remove the first 90% of moisture. Drying behavior of some biological and most food materials experience this second falling rate period.[9,27,30,34] When vapor pressure of the material becomes equal to the partial vapor pressure of the drying air, no further drying takes place,[11] and the moisture content at this stage is called the equilibrium moisture content, M_e. As the constant-rate and falling-rate periods are described, one can see that the material's water binding and porous structure influence the mechanism of moisture transfer.

The internal transfer mechanisms during convective hot air drying are heat conduction/convection from the surface to the center and the simultaneous transport of moisture as bulk liquid or as vapor from the material to unsaturated air.[9] External resistances to mass transfer become important for a Biot number for mass transfer, $Bi_m = k^*L/D\rho_s$ less than 10.[35] In addition, experimental studies of the effect of air velocity on the moisture content of the material during drying can be used to determine whether external resistances is important to mass transfer.[35] When convective hot air is equal to or greater than 1 m/s, external resistances become negligible.[34] Moisture loss during the falling rate period is an internally controlled process, and the moisture transfer mechanisms listed in well-referenced review in drying are given in Table 1. Much debate in developing an accurate model to predict drying behavior has been over which mode or modes of moisture transfer governs moisture loss and whether or not heat transfer effects should be taken into consideration. The difficulty arises in complex systems where several mechanisms can account for moisture transfer as drying proceeds due to a change in the system, though only one mechanism predominates moisture transfer at any given time.[9]

Table 1 Internal moisture transfer mechanisms.

Sherwood and Lewis in the 1920's.[38,39,40]
   Diffusion

Cegliske and Hougen in 1937.[41]
   Capillary

Henry in 1948.[105]
   Evaporation-condensation

Görling in 1958.[150]

Transfer of liquid water	**Transfer of water vapor**
Capillary flow	Differences in partial pressure (diffusion)
Liquid diffusion	Differences in total pressure (hydraulic flow)
Surface diffusion	

Keey in 1970:[151]
   Hydraulic flow
   Capillary flow
   Evaporation-condensation
   Vapor diffusion

Bruin and Luyben in 1980.[104]
   Molecular diffusion
   Capillary flow
   Knudsen flow
   Hydrodynamic flow
   Surface diffusion

Hallstöm in 1990.[152]

Transfer of liquid water	**Transfer of water vapor**
Molecular Diffusion (within solid)	Diffusion (in pores): Knudsen, ordinary, or Stephan diffusion.
Capillary flow (saturated capillaries)	Hydraulic flow (in pores)
Liquid diffusion (in pores)	Evaporation-condensation
Surface diffusion (absorbed water)	
Hydraulic flow (in pores)	

Waananen, Litchield, and Okos in 1993.[69]

Transfer of liquid water	**Transfer of water vapor**
Diffusion	Mutual diffusion
Capillary flow	Knudsen diffusion
Surface diffusion	Effusion
Hydrodynamic (bulk) flow	Slip flow
	Hydrodynamic (bulk) flow
	Stephan diffusion
	Poiseuille flow
	Evaporation-condensation

Diffusion

Diffusion is the process by which mass is transported from one part of a system to another part as a result of a concentration gradient. The process leads to an equilibrium of concentration within the system.[36] Simple examples of diffusion of mass are: liquid water evaporates into still air because of a difference in concentration between water vapor at the surface and air; a piece of sugar added to a cup of coffee dissolves by itself and diffuses to the surrounding solution; and odors diffuse in air.[8] Diffusion in liquids

and solids is a significant mechanism of mass transport in many chemical and biological pro-
cesses. Examples of food industrial processes involving diffusion in liquids are liquid-liquid
extraction for refining vegetable oils, extraction of caffeine in aqueous solutions, and drying of
liquid solutions, such as fruit juice, tea and coffee, to remove water, volatile flavor and aroma
compounds.[13] Diffusion in solids is also important in chemical and biological processing, as
shown in the following examples: liquid/solid extraction of sugars from beets or oils from
seeds and beans; drying of timber and foods; diffusion and catalytic reaction in solid catalysts;
separation of fluids in membranes; and diffusion of gases and moisture through polymer films
used in packaging.[8,13] Diffusion has been shown to occur widely in various industries to vari-
ous systems, and it is important to describe how this mechanism has been quantified.

The transfer of heat is a result of random molecular motion caused by a temperature
gradient. Fick recognized the similarity between heat transfer and mass transfer and was
the first to quantify diffusion by adopting the Fourier equation for heat conduction. The
diffusion flux, J_m, can be expressed by Fick's first law of diffusion:

$$J_m = -D\frac{\partial C}{\partial x},\tag{1}$$

where the rate of flow of a solute through a plane will be proportional to the cross-
sectional area, A; and the concentration gradient, $-\partial C/\partial x$. The flow of the solute from time
t to $t + \partial t$ can be expressed by the net flux change of diffusing molecules from high con-
centration to low concentration in a layer of fluid bounded by two parallel planes situated
at x and $x + \partial x$ as follows:

$$\frac{\partial C}{\partial t} = \frac{1}{\partial x}[J(x) - J(x+\partial x)] = -\frac{\partial J}{\partial x}.\tag{2}$$

Substituting the mass flux term expressed in Eqs. (1) into (2) results in Fick's sec-
ond law of diffusion:

$$\frac{\partial C}{\partial t} = \frac{\partial}{\partial x}\left(D\frac{\partial C}{\partial x}\right).\tag{3}$$

The potential for diffusion is a concentration gradient analogous to a temperature
gradient as the potential for heat transfer. Table 2 shows the mathematical similarities
between these two transports when D and k_T are constant.

Table 2 Heat and mass transfer equations.

Heat transfer	Mass transfer
$J_h = -k_T\dfrac{\partial T}{\partial x}$	$J_m = -D\dfrac{\partial C}{\partial x}$
$\dfrac{\partial T}{\partial t} = \left(\dfrac{k_T}{\rho c_p}\right)\dfrac{\partial^2 T}{\partial x^2}$	$\dfrac{\partial C}{\partial t} = D\dfrac{\partial^2 C}{\partial x^2}$

The diffusion equations have been attractive to use since analytical solutions for the differential equations describing temperature and concentration distributions within a body at any time are available provided that the physical properties of the material are constant.

Although the analogy between each transport is the same mathematically, the actual physical mechanisms are different. In heat transfer by conduction, the molecules of the medium are stationary and the transport is done mainly by electrons.[37] In mass transfer, one or more components are often being transported by relative motion through one another. The process by which matter is transported from one part of a system to another part is a result of random molecular motion, also known as random walk.[8] The cause of this molecular motion is a concentration gradient where the net flux of the diffusing substance is from a region of high concentration to a region of low concentration. In the diffusion of gases or the diffusion of a solute in a dilute solution, the molecules of the diffusing substance rarely collide with each other due to the tremendous spacing among the diffusing molecules. The resistance of diffusion comes from the collisions of the diffusing molecule with the molecules of the medium.[8] When diffusion occurs in higher concentrated solutions or in solids, the molecules of both the diffusing substance and the medium are closer together. The resistance of diffusion then is a result of collisions of the diffusing molecules with each other, as well as with the molecules of the medium. The diffusion coefficient becomes dependent on the concentration, and the diffusion equation takes the form represented in Eq. (3). The solution of this form of the diffusion equation becomes more complex. Crank[37] provides detailed solutions of the diffusion equation when the diffusivity is considered constant or concentration dependent for many initial and boundary conditions.

In 1921, Lewis[38] first recognized that diffusion could be applied in the dehydration process. Later, Sherwood[39] described a method to apply Fick's second law of diffusion to quantify moisture loss during the falling rate period in drying. Fick's Law was used to predict drying times for materials such as clay, wood, and soap, and the results were reasonably well.[39,40] Although drying rate and drying time predictions were fairly accurate, the diffusion equations did not predict moisture distributions within the materials.[41] The researchers pointed out the integration method used to solve the diffusion equation, in which the average moisture content was evaluated, corrected for any discrepancies between experimental data and mathematical predictions of moisture distribution and thus accurately predicted drying time. Hougen et al.[42] critically evaluated the diffusion equation and concluded on the following conditions when the diffusion could be properly applied:

1. single-phase solid systems in which water and the solid are mutually soluble, such as soap, gelatin, and glue;
2. wood and similar material solids below the fiber-saturation point; and
3. last stages of drying starches, textiles, paper, clay, hydrophilic solids, and other materials when bound water is being removed.

Food materials can be classified under the third condition as a hydrophilic or hygroscopic material which losses moisture in the bound water region or sorption region. Thus, moisture transfer in the falling rate period of drying can usually be described by a diffusion model based on Fick's second law.[43] However, Hougen et al.[42] pointed out that although Fick's law predicted drying time and drying rate for materials such as soap, clay,

and wood pulp fairly well, the diffusion equations did not provide accurate predictions for the moisture distribution under uniform drying conditions. The discrepancy between experimental data and predictions of moisture distribution in potato slices during drying was also observed by Ede and Hales.[44] Sherwood[40,45] reported that diffusion coefficient estimated from experimental data on drying of soap was not constant but decreased noticeably at low levels of moisture contents. This indicates that the better predictions of moisture distribution can be obtained if the concentration dependent diffusivity were used. Furthermore, for a system with complex network or structure, other mechanism(s) other than liquid diffusion may be responsible for the moisture movement in solids and should be taken into account to obtain more accurate drying model. Therefore, as the system becomes more complex, the further the diffusion equation deviates from the heat conduction equation in physical meaning and more complex solutions are required.

Despite the debate on which mechanism(s) contribute to the moisture transfer, liquid diffusion has been widely considered to be the rate-limiting mechanism responsible for internal moisture transfer in drying of many foods during the falling-rate period, such as wheat kernel,[46] potato starch gel,[47] fish muscle,[34] potato[28,48,49,50], tapioca root,[32] sugar beet root,[30] avocado,[31] rough rice grain,[51,52] bananas,[53] dense pasta,[17] green peas,[5] garlic,[54] plantain,[55] and corn.[56]

Many researchers have suggested that vapor diffusion as the mechanism by which moisture is transferred during drying of hygroscopic porous materials.[9,27,57–59] Van Arsdel et al.[10] claimed that better predictions of moisture distribution for drying of porous materials could be obtained if Fick's second law equation is stated in terms of vapor pressure gradient as the driving force for diffusion rather than moisture concentration gradient:

$$\frac{\partial p}{\partial t} = \frac{\partial}{\partial x}\left(P_D \frac{\partial p}{\partial x}\right), \tag{4}$$

where p is moisture vapor pressure, which is related to moisture content through a sorption isotherm; and P_D is the moisture dependent vapor diffusion coefficient, which relates the rate of moisture movement to a vapor pressure gradient.

The effect of total pressure on the moisture diffusivity was investigated in materials with different porosity, both dense and porous pasta.[17] For porous pasta, with porosity of 0.26, the dependence of moisture diffusivity on total pressure was more significant than for dense pasta, with a porosity of 0.06. This indicates that vapor diffusion significantly contributes to internal moisture transfer during drying of porous solids. The results from this study also showed the importance of food structure in determining the mechanisms of moisture transport during drying.

Capillarity

Capillary flow is the movement of liquid through capillaries, interconnecting pores in a solid, or over the surface of a solid by molecular attraction between the liquid and the solid.[42] The attractive forces between the liquid molecules and the solid is called adhesion forces, and it is these adhesion forces which causes some liquids like water to move up a channel from a region of high concentration to a region of low concentration. Capillarity is observed in many situations: fluid transfer up a wick; water migration up through soil to the ground surface; as well as in many life processes, such as the transport of water

in plants from the roots to the leaves and in distribution of blood throughout an animals body.[60] The equation for capillary flow is given:[61]

$$\ln\left(\frac{M - M_\infty}{M_i - M_\infty}\right) = \frac{h_s(T - T_s)}{\rho_s L \lambda (M_i - M_\infty)} t.$$

(5)

Mathematically, the capillary equation is similar to the diffusion equation. This similarity has often led to incorrectly defining capillarity as capillary diffusion.[62] Though both transfers have a similar driving force, a concentration gradient, the mechanisms of flow are different.

Ceaglske and Hougen[41] suggested that capillary flow and not diffusion flow be responsible for water transport in course granular solids. In a related study, Hougen et al.,[42] provided additional examples of when the capillary equation was appropriate, such as sand, paint pigments and minerals, as well as for hygroscopic materials in which the moisture content was above the sorption region. Several researchers have supported this observation and modeled drying of porous materials based on multiple moisture mechanisms, suggesting that capillarity as the rate-limiting mechanism for moisture transfer at the beginning of drying and then vapor diffusion dominating the latter stages of drying.[63–66] Saravacos and Charm[28] investigated mechanisms of water transfer during drying of some fruits and vegetables by studying the effect of surface-active agents on their drying rates. The results showed that there was no effect of the surfactants on the drying rate of all samples during the first falling-rate period. This implied that the moisture is not transferred by the capillary mechanism during this period of drying. Thus, capillary movement has not often been used for modeling moisture transfer in food systems.

Hydraulic Flow

Hydraulic flow is often used to describe the transfer of moisture through geological porous materials, such as to describe groundwater transfer in soil. In 1856, Henry Darcy reported his study on flow of water through beds of sand. The relationship Darcy presented is shown by the following one-dimensional flow:[67]

$$\frac{dQ}{dt} = KA\frac{\Delta h}{L},$$

(6)

where dQ/dt is the volumetric flow rate of a fluid (m^3/s); K is the hydraulic conductivity (m/s); A is the cross-sectional area to flow (m^2); L is the flow path length (m); and Δh is the change in hydraulic head over path L (m). The hydraulic head, h, at a specific point is the sum of the pressure head and elevation or gravitational head:

$$h = (p/\rho g + z),$$

(7)

where p is the water pressure (N/m^2); ρ is the water density (kg/m^3); g is the acceleration of gravity (m/s^2); and z is the elevation head (m). Substituting Eq. (7) into Eq. (6),

$$\frac{dQ}{dt} = KA\frac{\Delta(p/\rho g + z)}{L}$$

(8)

and is a similar form Darcy used to analyze his experiments. The relationship of hydraulic conductivity and permeability is shown

$$K = \frac{k_p \rho g}{\mu}, \tag{9}$$

where k_p is the permeability (m^2); and μ is the fluid absolute viscosity (N s/m^2). Under unsaturated conditions, the elevation head is negligible compared to the pressure head,[68] and substituting Eq. (9) into Eq. (8) and cancelling the density and gravity terms, the resulting expression is a common form of Darcy's law:

$$\frac{dQ}{dt} = \frac{k_p A}{\mu} \frac{\Delta p}{L}. \tag{10}$$

Like Fourier and Fick's laws, Darcy's law is a phenomenological expression; however, Darcy's law has been derived from Navier-Stokes equations. Though hydraulic flow is often used to describe groundwater transport, few have applied this to moisture transfer in foods. Pressure driven flow may be applicable to high temperature drying near boiling or vacuum drying where evaporation rates are significant.[68,69,72]

Evaporation-Condensation

Evaporation-condensation can be considered as a coupled internal heat and mass transfer mechanism where mass is transferred in the vapor state and heat is gained and lost by condensation and evaporation, respectively. Harmathy[73] examined the heat and mass transfer of a porous system during drying and concluded that evaporation-condensation was the dominate mechanism during falling rate drying. With appropriate initial and boundary conditions, a set of second order partial differential equations were used to obtain complete moisture, temperature and pressure distributions throughout the drying of a brick slab.[73] Harmathy concluded that the discrepancies with the model predictions to experimental data were attributed to varying surrounding temperature and relative humidity, as well as the assumption of constant latent heat of vaporization and sorption which were also assumed to be equal.

Further evidence of evaporation and re-condensation as the governing mechanism for hygroscopic porous materials were observed from drying studies on bread samples. The diffusion model based on Fick's second law was unable to describe the drying characteristics of bread during microwave heating,[74] during convective hot air drying,[75] and during isothermal drying.[76] Louvet[77] studied the drying rates of pre-gelatinized dough samples during microwave heating at different power levels with and without the presence of saturated steam at the surface. No significant difference in drying rate was observed between drying conditions with or without steam and when the other drying parameters, such as temperature history, sample size and microwave power, were kept constant. Since the drying rate would have been affected by the drying conditions if the drying were diffusion limited, another mechanism other than diffusion would dictate the drying of bread. Also, the drying rate was found

to increase with increasing microwave power even when the temperature histories of the samples were quite similar. Thus, the authors further hypothesized a drying mechanism limited by the availability of enthalpy of vaporization governs the moisture loss of bread during drying. Tong et al.[78] measured the pressure profiles in bread samples as a function of sample porosity during microwave heating. The results showed that pressure gradients were negligible when the porosity was greater than 0.3, which also suggests that the rate of vaporization of water is less than the rate of vapor migration and therefore is vaporization limited. This observation follows the research on permeability of bread by Goedeken and Tong,[16] where permeability significantly increased above porosity 0.3. As pointed out by Waananen et al.,[69] this negligible pressure gradient was also observed in iron ore pellets and silica samples with porosities above 0.3 (Cross et al.;[79] Gibson et al.[80]). Tong et al.[81] observed uniform moisture profiles in bread during isothermal drying at 100°C established using microwave energy and convective hot air, as shown in Figure 1. Since a diffusion controlling mechanism would have resulted in a parabolic moisture profile,[30,82] the researched concluded
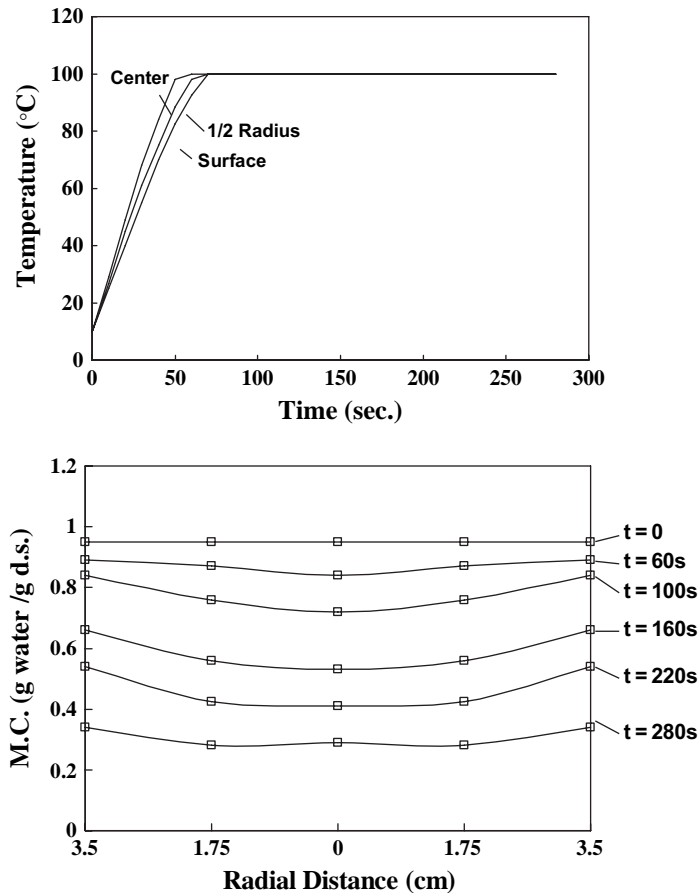
Figure 1 Isothermal drying of bread at 100°C: a) temperature profile; b) moisture profile. Adapted from Tong et al.[81]

that isothermal drying of bread is not diffusion limited. Litchfield[83] concluded that high rates of mass flux results in flatter moisture profiles. Thus, it can be concluded that such high mass flux rates suggests mass transfer may not be governed by diffusion but rather a heat transfer limited phase-change mechanism, such as evaporation-condensation. In analyzing the temperature profiles of bread and dough samples during convective hot air drying, constant temperature was observed at the center of bread[75,84,85] and dough[86] samples. These center temperatures remained relatively constant throughout for most of the drying time. Constant center temperature profiles during drying of wool bobbin was also observed and termed pseudo-wet-bulb temperature.[87]

Sluimer and Krist-Spit[84] showed that when porous sample temperatures are below 100°C and an established temperature gradient throughout, and vapor generated in the interior regions between the surface and center may diffuse towards the cooler center region and re-condense. This recondensed vapor releases latent heat to the material, which is consistent with observed temperature profiles of the dough and bread samples during convective hot air drying where a constant temperature was observed at the center of the bread samples followed by a decrease in temperature and then followed by an increase toward the drying temperature.[75,86] The pseudo wet bulb temperature observed may have been as result of recondensation, the decrease in temperature may have been due to evaporation, and then the final increase in temperature towards the dry-bulb temperature due to drying of the center region. Thorvaldsson and Janestad[85] also showed an increase in moisture at the center while center temperatures remain constant at a pseudo-wet-bulb temperature during dehydration of bread at 210°C and concluded evaporation-condensation as the governing mechanism for dehydration.

The significance of the pseudo-wet-bulb temperature is shown in the following energy balance:

$$\rho c_p \frac{\partial T}{\partial t} = k_T \frac{\partial^2 T}{\partial x^2} + \lambda \rho_s \frac{\partial M}{\partial t}. \tag{11}$$

For the center region experiencing pseudo-wet-bulb temperature, the left-hand side of Eq. (11) is zero:

$$0 = k_T \frac{\partial^2 T}{\partial x^2} + \lambda \rho_s \frac{\partial M}{\partial t}. \tag{12}$$

As Eq. (12) describes, as well as been observed by the pseudo-wet-bulb temperature region, the rate of heat conduction to the center is equal to the rate of heat evolved during condensation or equal to the rate of evaporative cooling in this region. Heat is gained as moisture absorbs into the material, or heat is lost as moisture evaporates from the material. When a constant temperature is observed, such as the case with this pseudo-wet-bulb temperature profile, a change of phase is occurring within the material and suggests that available latent heat of vaporization is limiting the mechanism of moisture loss during drying; thus, heat transfer cannot be neglected.

SIMULTANEOUS HEAT AND MASS TRANSFER (SHMT) MODELS

Heat and mass transfer must be considered simultaneously in order to best describe the drying process. In order to remove moisture from a material, energy must be supplied for the evaporation of the water. In many systems, heat transfer can slow mass transfer rates substantially, and can even control the rate of moisture removal completely.[88] In addition, Luikov[64] showed that moisture transport inside a material may result from a significant temperature gradient (thermogradient effect). For these reasons, heat transfer effect should be taken into account along with mass transfer when developing a drying model.

Modeling the transport mechanism of drying process involves setting up the heat and mass transfer equations correctly, solving the coupled differential equations, and then correlating the prediction with experimental moisture and temperature profiles.[19] In general, drying models can be classified into 3 groups: 1) those involving empirical equations applicable for specific processes, 2) those based on simultaneous equation systems of basic heat and mass diffusion models, and 3) those associating energy, mass and momentum transport equations with all the thermodynamically interactive fluxes.[89] The last group of simultaneous heat and mass transfer models may involve more than one mechanism of mass transport and thus is more sophisticated. These complicated models have been derived based on mechanistic approaches[63] or from non-equilibrium thermodynamics.[64,90]

Two useful methods used to solve a set of differential equations in order to describe drying behavior in regular shaped bodies are analytical methods (i.e., the variable separation method) and numerical methods (i.e., the finite element and difference method).[91] Most models are based on assumptions of movement, structure and thermodynamics for the purpose of simplifying the heat and mass transfer equations. With numerical approaches, many simplifying assumptions can be eliminated.[92]

Many researchers have developed mathematical models to describe drying processes in foodstuffs.[10,27,93] There are generally three types of approaches for developing drying models: 1) the models based on the concept of characteristic drying curves in different drying stages; 2) the distributed-parameter models, using coupled heat and mass diffusion equations; and 3) the empirical models obtained entirely by simple or multivariable regression methods.[94] Several types of models are commonly used to describe the falling rate period of drying. These include the diffusion model which is based on the diffusional transport of water, the receding front model which is based mostly on capillary transport, and the model based on the complete conservation equations, which give mathematically complex formulations.[95] The levels of model complexity depend on consideration of shrinkage[96] or no shrinkage,[97] assuming isothermal conditions[2] or non-isothermal conditions,[98] and assuming moisture concentration dependence of the diffusion coefficient.[99,100] Two major boundary conditions distinguishing these models are moving boundary condition, which takes shrinkage into account, and equilibrium at the surface, which is associated with no external resistance. Most models are usually based on the assumptions that the external surface of the material is at equilibrium and the geometry (shape) is unchanged (no shrinkage).[95,101]

Many simultaneous heat and mass transfer models have been developed for drying based on either lumping the moisture transfers to one effective diffusivity or separating liquid diffusion and vapor diffusion. A brief review of these models is presented along with underlying assumptions to simplify the models.

SHMT Using Effective Diffusivity

King[59] used Fick's second law to model heat and mass transfer in a hygroscopic porous body, and used an "effective diffusivity" term, D_{eff}, which lumps internal mass transfer mechanisms together, as shown below:

$$\frac{\partial M}{\partial t} = \frac{\partial}{\partial x}\left(D_{eff}\,\frac{\partial M}{\partial x}\right), \tag{13}$$

where, in the absence of internal temperature gradients, the effective diffusivity is:

$$D_{eff} = \frac{M_w D' P_w^o}{\rho_s R_v T}\left(\frac{P}{(P-p_w)}\right)\cdot\left(\frac{\partial a_w}{\partial M}\right)_T. \tag{14}$$

In this equation, the effective diffusivity relates the moisture content to the vapor pressure, which is the driving force for diffusion. The function of the term $(\partial a_w/\partial M)_T$ can be linear or non-linear, depending on the range of moisture constant, and determines whether the effective diffusion coefficient can be considered constant or dependent on moisture content. If the relation between vapor pressure and moisture content is not linear but curved as seen in most food isotherms, the term $(\partial a_w/\partial M)_T$ is not constant and D_{eff} will vary with moisture content. However, drying models based on the mechanism of vapor diffusion for some porous food materials, such as peanut pods,[102] and grain sorghum,[103] have been developed by assuming constant effective diffusion coefficient.

King[59] further pointed out that the desorption of a material requires heat to be consumed to vaporize the moisture, thus making the drying of hygroscopic porous materials as an interaction of heat and mass transfer. The effective diffusivity term can also be expressed to include internal heat transfer, as shown below:

$$D_{eff} = \frac{M_w D' P_w^o}{\rho_s R_v T}\left(\frac{P}{(P-p_w)}\right)\cdot\left(\frac{\partial a_w}{\partial M}\right)_T\cdot\left(\frac{\psi}{1+\psi}\right), \tag{15}$$

where,

$$\psi = \frac{k_T R R_v T^3}{(\Delta H_d)^2 D' a_w P_w^o} \tag{15b}$$

The major assumptions upon which this model was based are: 1) mass transfer is by vapor diffusion; 2) a local equilibrium exists between the vapor phase and the sorbed moisture; 3) the heat of desorption is considerably greater than sensible heat of the medium; 4) the amount of sorbed moisture is large in comparison to the water vapor; therefore, as the vapor diffuses out of the material it is constantly replenished by evaporation of the sorbed moisture; 5) the heat of desorption is constant; and 6) shrinkage is negligible.

Equation (15) describes either a heat transfer controlled or a mass transfer controlled process. The term $\{\psi / (1 + \psi)\}$ expresses the limiting transfer mechanism and can be represented by the following:[62]

$$\frac{\psi}{1+\psi} = \frac{k_T RR_v T^3}{k_T RR_v T^3 + (\Delta H_d)^2 D' a_w P_w^o} = \frac{\text{heat transfer}}{\text{heat transfer} + \text{heat transfer by mass transfer}}.$$

If the thermal conductivity, k_T, is large compared to effective vapor diffusivity, D', then $\psi \gg 1$ and $\{\psi / (1+\psi)\}$ become unity. The effective diffusivity is then represented by equation (14) which shows a direct relationship to D'. If the effective vapor diffusivity is significantly greater than the thermal conductivity, then $\psi \ll 1$ and $\{\psi / (1+\psi)\}$ is equal to ψ, and heat conduction controls the drying process. The advantage of this model is the ease in which a drying process can be evaluated based on fundamental transport properties in the effective diffusivity expression.[93]

More complicated diffusion theory can be obtained by taking Fick's law as representative for both liquid and vapor transfer.[1] Waananen and Okos[17] defined a total effective diffusivity as accounting for liquid and vapor fluxes:

$$D_{eff} \approx \frac{C_2'}{P} + C_1', \tag{16}$$

where C_1' and C_2' are parameters characterizing liquid and vapor contributions, respectively. For vapor diffusion, D_{eff} is assumed to be proportional to total pressure, so the term $1/P$ is incorporated with the vapor contribution parameter (C_2').

SHMT Using Separate Liquid and Vapor Fluxes

Several researchers have developed simultaneous heat and mass transfer models based on the assumption that moisture transfer occurs both in the liquid and vapor state. Luikov[64] developed a model for simultaneous heat and mass transfer in capillary porous materials based on irreversible thermodynamics which takes into account all thermodynamic driving forces. Of particular interest, Luikov[64] introduced the concept of moisture transfer caused by a temperature gradient in addition to a concentration gradient. The liquid and vapor fluxes, J_{liq} and J_{vap} respectively, are expressed showing the two driving forces for transfer:

$$J_{liq} = -D_{liq}\rho_s \left[\frac{\partial M}{\partial x} + \delta_{liq} \frac{\partial T}{\partial x} \right], \quad \text{and} \tag{17}$$

$$J_{vap} = -D_{vap}\rho_s \left[\frac{\partial M}{\partial x} + \delta_{vap} \frac{\partial T}{\partial x} \right], \tag{18}$$

where D represents the mass transfer coefficient; and δ represents the thermal gradient coefficient. The major assumptions in Luikov's model are: 1) moisture is transfer out of the system by liquid and vapor diffusion simultaneously; 2) the thermal and diffusion

properties are constant; and 3) the total pressure is constant. Thus, the differential heat and mass transfer equations are shown:

$$\frac{\partial T}{\partial t} = \alpha \frac{\partial^2 T}{\partial x^2} + \in \frac{\lambda}{c_p} \frac{\partial M}{\partial t}, \quad \text{and} \tag{19}$$

$$\frac{\partial M}{\partial t} = (D_{liq} + D_{vap}) \frac{\partial^2 M}{\partial x^2} + (D_{liq} + D_{vap}) \delta \frac{\partial^2 T}{\partial x^2}. \tag{20}$$

The phase conversion factor, $\in$, is a dimensionless parameter expressing the degree of evaporation within the material. Vapor diffusion is expressed when $\in = 1$, thus liquid diffusion would be absent and a change in moisture content of the material would occur due to evaporation of the liquid or condensation of vapor. The phase conversion factor is zero when liquid diffusion is the primary mode of transfer, thus evaporation within the material is negligible.[64]

Analytical solutions to Luikov's model has been widely studied. However as pointed out by Rossen and Hayakawa,[93] the major limitation of the model comes from the assumption of constant thermophysical propteries. If the thermophysical properties are expressed as functions of moisture content and temperature, the solution to Luikov's model would become exceedingly complex.

Similarly but independently, Philip and De Vries[63] developed a simultaneous heat and mass transfer model for porous materials with driving forces of moisture and temperature gradients. Philip and DeVries[63] used classical mechanisms of vapor diffusion and liquid movement by capillarity to show the influence of temperature gradient has on moisture movement. These authors started with the modified theory of vapor diffusion as applied to porous material and the separation of isothermal vapor transfer and thermal vapor transfer:

$$J_{vap} = -D_{Tvap} \frac{\partial T}{\partial x} - D_{\theta vap} \frac{\partial M}{\partial x}, \tag{21}$$

where D_{Tvap} and $D_{\theta vap}$ are the thermal vapor diffusivity and isothermal vapor diffusivity, respectively. The liquid flux was based on Darcy's law for liquid transfer in unsaturated porous material and separated into isothermal liquid transfer and thermal liquid transfer:

$$J_{liq} = -D_{Tliq} \frac{\partial T}{\partial x} T - D_{\theta liq} \frac{\partial M}{\partial x} - J_g, \tag{22}$$

where D_{Tliq} and $D_{\theta liq}$ are the thermal liquid diffusivity, and isothermal liquid diffusivity, respectively; and J_g represents the liquid flux due to gravity. The authors then combined the vapor flux equation, Eq. (21), with the liquid flux equation, Eq. (22) into one overall mass transfer equation:

$$J = -(D_{Tliq} + D_{Tvap}) \frac{\partial T}{\partial x} - (D_{\theta liq} + D_{\theta vap}) \frac{\partial M}{\partial x} - J_g. \tag{23}$$

Equation (23) can be reduced to:

$$J = -D_T \frac{\partial T}{\partial x} - D_\theta \frac{\partial M}{\partial x} - J_g, \qquad (24)$$

where, D_T = overall thermal moisture diffusivity = $D_{Tliq} + D_{Tvap}$, D_θ = overall isothermal moisture diffusivity = $D_{\theta liq} + D_{\theta vap}$. The general differential equation describing heat and moisture transfer in porous material with temperature and moisture gradients are expressed:

$$c_v \frac{\partial T}{\partial t} = \frac{\partial}{\partial x}\left(k_T \frac{\partial T}{\partial x}\right) - \lambda \frac{\partial}{\partial x}\left(D_{\theta vap} \frac{\partial M}{\partial x}\right), \quad \text{and} \qquad (25)$$

$$\frac{\partial M}{\partial t} = \frac{\partial}{\partial x}\left(D_T \frac{\partial T}{\partial x}\right) + \frac{\partial}{\partial x}\left(D_\theta \frac{\partial M}{\partial x}\right) + \frac{\partial J_g}{\partial z}. \qquad (26)$$

At high moisture contents, the two liquid diffusivities, D_{Tliq} and $D_{\theta liq}$, are important, and at low moisture contents, the two vapor diffusivities, D_{Tvap} and $D_{\theta vap}$, are important. As pointed out by Rossen and Hayakawa,[93] a major disadvantage in using Philip and De Vries' model was its complexity with four moisture-dependent diffusivities.

Berger and Pei[65] developed a simultaneous heat and mass transfer model based on theories from Krischer, which also assumed mass transfer occurring by liquid capillary flow and vapor diffusion. The major criticism of Krischer's model was in using sorption isotherm as the coupling equation throughout the entire moisture range to predict drying behavior. Berger and Pei[65] used the Clausius-Clapeyron equation as the coupling equation in the maximum sorption range and the sorption isotherm as the coupling equation in the sorption region. However, their model has not been widely used to predict drying behavior of biological materials.[1]

Whitaker[66] developed a simplified theory that consists of a set of coupled, volume-averaged transport equations for the temperature and moisture content. The theory incorporates liquid and vapor phase continuity equations, combines liquid, solid and vapor phase thermal energy equations into a single temperature equation and makes use of Darcy's law for liquid and vapor phases to account for moisture transport. As pointed out by Bruin and Luyben[104] the difficulty in this approach is in justifying the validity of the local equilibrium assumption among the three phases, though Datta and colleagues have stated that they are working on a non-equilibrium approach to this problem.[70]

Evaporation-Condensation

Henry[105] developed a model to describe the coupled mechanisms of the heat absorbed when water evaporates from a solid and the heat evolved when water vapor

condenses in a solid. Thus, Henry's model is commonly referred to as the evaporation-condensation model.[73] The energy and mass balances are given:[42]

$$\frac{\partial T}{\partial t} = \alpha \frac{\partial^2 T}{\partial x^2} + \frac{\lambda}{c_p} \frac{\partial M}{\partial t}, \quad \text{and} \tag{27}$$

$$\frac{\partial C}{\partial t} = D_{AB} \frac{\partial^2 C}{\partial x^2} - \frac{\rho_s (1-\Phi)}{\Phi} \frac{\partial M}{\partial t}. \tag{28}$$

The assumptions of this model are: 1) the moisture concentration, water vapor concentration and temperature can be expressed in a linear moisture sorption isotherm equation:

$$M = \text{constant} + \sigma C - \omega T, \tag{29}$$

where σ and ω are constants; 2) the parameters diffusion coefficient for moisture in air, over-all heat conductivity, specific heat and density are constant and independent of moisture concentration and temperature; 3) the latent heat is constant and equal to that of free water; 4) no hysteresis effect; 5) porosity is constant; 6) moisture diffusion takes place only in the vapor phase; and 7) a local equilibrium exists between the solid and pore spaces.

The limiting assumption of this model is the first assumption, the linear relationship of vapor concentration to moisture concentration and temperature, which rarely exists in food materials. However, the relationship between moisture content and vapor concentration can be shown in a moisture sorption isotherm, of which numerous equations have been offered. It is well known that the equilibrium moisture content to water activity relationship has a sigmoidal shape.[9,11] The most common mathematical models to predict moisture sorption isotherms along with their most common application are widely given in literature.[11,62] The GAB model is reported to be the best model to predict moisture sorption isotherms over a wider range of water activity, $0 < a_w < 0.9$, for many food products.

Whitney and Porterfield[106] modified Henry's model by including an internal heat generation term, Q, in the energy balance equation when the drying process involve dielectric heating:

$$\rho_s c_p \frac{\partial T}{\partial t} = k_T \frac{\partial^2 T}{\partial x^2} + \lambda \rho_s \frac{\partial M}{\partial t} + Q. \tag{30}$$

These authors tested Henry's model (without internal heat generation) and the modified model (with internal heat generation) on corn meal, and the predicted results agreed reasonably well with experimental data.[106]

Thorvaldsson and Janestad[85] developed an evaporation-condensation model using heat transfer, liquid diffusion, vapor diffusion, and the three equations were coupled by the saturated partial water vapor pressure. The predicted moisture profiles were reasonably

close to experimental profiles; however, the shortcoming of this approach is the assumption of local equilibrium between temperature, water vapor and moisture content.

Roberts and Tong[76] developed an isothermal drying apparatus to study drying behavior of foods at temperatures between 40–70°C and to quantify moisture transfer properties under isothermal conditions. During isothermal drying where no temperature gradients exist and thus no re-condensation, Roberts and Tong[107] proposed that internal evaporation would be the rate limiting mechanism for isothermal drying of hygroscopic porous materials and considered a reaction kinetic approach to model evaporation through a porous food material as illustrated below:

$$\text{M} \xrightarrow{\ k\ } \text{V}$$
$$\text{(Moisture within Sample)} \qquad \text{(Water Vapor)}$$
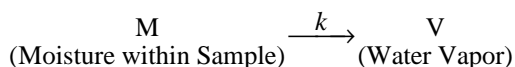
Roberts and Tong[107] showed that this irreversible first order kinetic model predicted moisture profile loss during isothermal drying of bread throughout the entire moisture range from 0.9 to 0.002 g/g dry solid. It was interesting to observe that the activation energy measured for the evaporation rate constant was slightly greater than latent heat of evaporation of pure water.

The limitation of an irreversible first-order reaction kinetic approach is that it can only be applied, mechanistically, to the case where a porous material is being dried isothermally along with negligible resistance to external moisture transfer. The moisture transfer is much more complicated under non-isothermal conditions, such as during convective hot-air dehydration where a temperature gradient is established. Under isothermal conditions, once internal moisture evaporates it diffuses to the dry convective air due to the partial vapor pressure gradient. When non-isothermal conditions are established, the water vapor in a pore can diffuse in two directions: 1) towards the drier convective air; and 2) towards the cooler center region. Water vapor diffusing in this cooler center region could condense or at least increase the vapor pressure in the pore space.

Dehydration of hygroscopic mid and highly porous ($\Phi > 0.25$) materials appear to be governed by phase change, which is heat transfer limited, and would certainly explain the observed uniform moisture profile decrease during isothermal drying as well as explain the increase in moisture in center regions during convective hot air drying. Developing models, particularly mechanistic models, to describe this evaporation-condensation will continue to be a demanding challenge in the future.

EXPERIMENTALLY MEASURING EFFECTIVE MOISTURE DIFFUSIVITY

Experimental methods for measuring the effective diffusivity include drying curves, sorption kinetics, permeation methods, nuclear magnetic resonance (NMR), and radiotracer methods.[37,62,108] Saravacos and Maroulis[109] state that due to the complexity of moisture transfer, the measurement of effective diffusivity should be similar to the application of interest. Sorption kinetics and permeation methods are commonly used to quantify moisture transfer during food storage, and an excellent review of these methods are provided by Saravacos and Maroulis.[109] Thus to quantify moisture transfer during drying, the effective moisture diffusivity should be measured from drying curves or from moisture profiles obtained during drying. The most common method of measurement is by

drying curves, and because of it popularity and sometimes misconception, this review of effective diffusivity measurement focuses on drying curve method and important assumptions underlying this method that are sometimes validated and often violated. Moisture profile measurements are important to determine the transfer mechanisms, to quantify moisture transfer parameters, and to provide experimental data to test model predictions, thus the accuracy of such a measurement is apparent. Of course it is very important that such measurements are conducted under the application of which the moisture transfer is studied. The second part of this section will cover advanced methods of moisture profile measurements, such as NMR.

Drying Curve Method of Measurement

Certainly there are limitations and over-simplifications in models using effective moisture diffusivity, but currently it is the most widely used equation to model dehydration. Saravacos and Maroulis[109] have reported since 1975 over 1700 data on effective moisture diffusivity have been published for various food and agricultural materials. And the most widely used method to measure effective moisture diffusivity is experimentally from drying curves based on the solution of Fick's second law equation. Assuming a constant diffusion coefficient, Fick's equation with one-dimensional diffusion for different geometries (slab, cylinder and sphere) can be given as:[11]

$$\frac{\partial M}{\partial t} = D_{eff}\left(\frac{\partial^2 M}{\partial r^2} + \frac{\eta}{r}\frac{\partial M}{\partial r}\right), \tag{31}$$

where η is constant and is dependent on shape: 0 for planar, 1 for cylindrical and 2 for spherical geometries. The initial and boundary conditions are:

$$M(r,0) = M_i, \quad \text{at } t = 0$$
$$M(0,t) = M_\infty, \quad \text{at } r = r_0 \text{ (at the surface)}$$
$$M(0,t) = \text{finite}, \quad \text{at } r = 0 \text{ (at the center).}$$

Based on assumptions of a uniform initial moisture distribution, negligible external resistance, negligible temperature gradients and negligible shrinkage during drying, the analytical solutions of the diffusion equations for simple geometries are given in the form of infinite series:[37]

for an infinite slab:

$$M^* = \frac{M - M_\infty}{M_i - M_\infty} = \frac{8}{\pi^2}\sum_{n=0}^{\infty}\frac{1}{(2n+1)^2}\exp\left[-\frac{(2n+1)^2\pi^2}{4l^2}D_{eff}t\right]; \tag{32}$$

for an infinite cylinder:

$$M^* = \frac{M - M_\infty}{M_i - M_\infty} = \sum_{n=1}^{\infty}\frac{4}{r^2\alpha_n^2}\exp(-D_{eff}\alpha_n^2 t), J_o(r\alpha_n) = 0; \quad \text{and} \tag{33}$$

for a sphere:

$$M^* = \frac{M - M_\infty}{M_i - M_\infty} = \frac{6}{\pi^2} \sum_{n=1}^{\infty} \frac{1}{n^2} \exp\left[-\frac{n^2 \pi^2}{r^2} D_{eff} t \right], \tag{34}$$

where M^* represents the unaccomplished moisture content; $J_o(x)$ in Eq. (33) is the Bessel function of the first kind and zero order; and α_n's are the roots of this function. Expansion of Eqs. (32), (33), and (34) for the first three series is shown:[110]

for an infinite slab:

$$M^* = \frac{8}{\pi^2}\left(e^{-\left(\frac{\pi}{2}\right)^2 N_{Fi}} + \frac{1}{9} e^{-9\left(\frac{\pi}{2}\right)^2 N_{Fi}} + \frac{1}{25} e^{-25\left(\frac{\pi}{2}\right)^2 N_{Fi}} \right); \tag{35}$$

for an infintie cylinder:

$$M^* = 0.692 e^{-5.78 N_{Fi}} + 0.131 e^{-30.5 N_{Fi}} + 0.0534 e^{-74.9 N_{Fi}}; \quad \text{and} \tag{36}$$

for a sphere:

$$M^* = 0.608 e^{-9.8 N_{Fi}} + 0.152 e^{-39.5 N_{Fi}} + 0.0676 e^{-88.8 N_{Fi}}, \tag{37}$$

where N_{Fi} is the Fick number $((D_{eff} t)/L^2)$; and L is the characteristic dimension of radius or half-thickness, depending on the geometry. For long drying times where the Fick number is greater than 0.1 and $M^* < 0.6$, the first term of the series dominates.[11,37,61] The equation is therefore simplified to the straight-line equation as follows:

$$\ln M^* = \text{Constant} - st, \tag{38}$$

where s is the slope and represents the dehydration constant (t^{-1}). The dehydration constant is also equal to $((c\, D_{eff})/L^2)$, where c is constant in the first series, as shown in Eqs. (35–37): for slab, $c = 2.47$; for cylinder, $c = 5.78$; for sphere, $c = 9.8$. Semi-log plot of unaccomplished moisture ratio and drying time represents a linear relationship above a Fick number $(D_{eff}t/L^2)$ of 0.1 and the effective moisture diffusivity can be calculated from the slope of the straight line using the following equation:[11,37,61]

$$D_{eff} = \frac{-s(L^2)}{c}. \tag{39}$$

The underlying assumptions required to accurately determine the diffusivity using the procedure outlined above are: 1) isothermal drying conditions; 2) constant effective diffusivity; 3) negligible shrinkage occurs during drying; 4) uniform initial moisture content; and 5) negligible external resistance.

Effect of Temperature on D_{eff} Measurements

Moisture loss during convective hot air drying is a coupled heat and moisture transfer mechanism, therefore, the assumption of negligible temperature gradients is the major assumption in using Fick's Second Law alone to analyze the drying curve and measure the effective moisture diffusivity. The assumption of negligible temperature gradients during experimental measurement of the effective diffusivity is the major assumption to use the diffusion equation. Measuring moisture transfer parameters within food materials becomes complicated when both heat and mass transfer processes have to be taken into consideration. King[59] analyzed theoretical conditions of when heat transfer effects should be considered and when they should not be considered. Heat transfer should be taken into account when the thermal conductivity is small and the diffusivity is large. Conversely, heat transfer does not need to be considered when the thermal conductivity of the material is large and the diffusivity is small.

Similar to this physical concept, Lewis number is used when a process involves simultaneous heat and mass transfer. Lewis number is the ratio of the thermal diffusivity $(\alpha_T = \dfrac{k_T}{\rho C_p})$ to the mass diffusivity (D): $Le \equiv \dfrac{k_T}{\rho C_p D}$ and represents the relative effect of temperature and moisture gradients inside the material. Young[88] agreed with King in that drying is a simultaneous heat and mass transfer problem, but Young[88] went a step further by quantifying when heat transfer effects should be considered. Young analyzed the effects thermal conductivity and diffusivity has on drying curves. Young[88] simplified Henry's model by introducing a modified Lewis number, Le_m, to indicate when heat transfer could be neglected:

$$Le_m = \frac{k_T\,[\Phi + (1-\Phi)\rho_s\beta]}{D_{eff}\,(1-\Phi)\rho_s\,(c_{ds} + c_w M + h\gamma)}. \tag{40}$$

The results showed that a modified Lewis number of 60 represented the minimum ratio where just the mass transfer alone described the drying curve. Below Le_m of 60, the heat and mass transfer equations have to be solved simultaneously to accurately predict drying behavior. It was reported that most biological materials have $Le_m > 60$,[88] and thus making drying of biological materials an internal mass transfer controlling process.

In another temperature gradient analysis, the internal temperature of food during drying can be considered uniform if Biot number for heat transfer, $Bi_h = (h_s L)/k_T$ is less than 0.1.[111] Alzamora et al.[112] calculated values of the Biot number for the various vegetables such as avocado, apple, potato and sugar beet root and found that they are in the range 0.2–0.5, and they concluded that there would be negligible temperature gradients during drying kinetic studies of foods.

Temperature measurements within different foods have been conducted by several researchers to experimentally verify negligible internal temperature gradients.[28,30,32,34,112] The negligible temperature gradients agree with the small Biot numbers and Lewis numbers over 60. However, these profiles also show that the sample temperatures varied with drying time and were well below the drying air temperature throughout most of the drying. In fact, the temperatures were not within 5°C of the drying temperature until over 90% of the moisture had been removed. Large temperature gradients were observed within the

bread samples with various porosities in sample diameters below 2 cm.[75,85] Wolf and Bimbenet[113] reported a 20°C temperature gradient between the center and surface of a carrot cubes during drying. Similar temperature gradients between the surface and center were observed during convective hot air drying of potato and carrot[114] and apple.[115]

Vaccarezza et al.[35] analyzed the effect of the changing sample temperature on the applicability of Fick's law. Based on assumptions of a uniform temperature throughout the solid and constant heat transfer coefficient, the sample temperature as a function of time was obtained using the macroscopic heat balance (Eq. 41) and the experimentally determined moisture content-time relationship (Eq. 42).

$$(m_w c_{pw} + m_s c_{ps}) \frac{dT}{dt} - h_s A(T_{db} - T) = \lambda \frac{dm_w}{dt}, \quad \text{and} \tag{41}$$

$$\frac{\bar{M}}{M_i} = \sigma \exp(-st) = \sigma \exp(-\pi^2 F_o), \tag{42}$$

where s and σ are the slope (sec^{-1}) and intercept, respectively, of the experimental straight line from ln $\bar{M}/M_i$ vs t. The knowledge of the sample temperature evolution obtained was then used to solve Fick's law with the temperature dependent D_{eff} (independent on moisture concentration). It was found that the application of Fick's law in predicting moisture profiles during the first falling rate drying of food materials such as sugar beet root was improved when the effect of the varying sample temperature was taken into consideration.

Alzamora et al.[112] also developed a simplified heat transfer to predict the temperature history during air-drying of vegetables. The model was derived using the energy balance with moisture evaporation at the surface and the moisture content-time relationship (Eq. 42). With only knowledge of the food drying curve and some physical properties, the model was found to give satisfactory predictions of temperature during air drying of avocado, potato, sugar beet root, and apple.

Moisture diffusivity of most food materials increases with the increase in drying temperature.[116] Generally, the temperature dependence of the effective diffusivity has been shown to follow an Arrhenius relationship in many studies:[28,30,34,103,117,118]

$$D_{eff} = D_o \exp\left(-\frac{E_a}{RT}\right). \tag{43}$$

An activation energy for diffusion, which can be determined from the slope of the Arrhenius plot ln (D_{eff}) vs. $1/T$, is typically less than 34 kJ/mol.[119] The temperature used in the Arrhenius analysis is the ambient temperature of drying, thus assuming that the temperature of the material being dried is that of the surrounding drying environment. Therefore, the isothermal assumption has been applied in both determining the moisture transfer parameter and in determining the activation energy.

However, few studies have actually obtained and verified isothermal conditions throughout drying when measuring diffusion coefficients, and though uniform temperature is usually assumed, many researchers have suggested that the discrepancies between the experimental data and model predictions result from the fact that the effective diffusion

coefficient is determined when sample temperature gradients exist.[27,35,59,119] Isothermal conditions may be possible during drying by convective hot air for either a non-porous or a porous material having a very small characteristic dimension. Waananen and Okos[17] measured center temperature for dense pasta (0.06 porosity) and porous pasta (0.26 porosity) during convective hot air drying at several temperatures. The center temperatures reached the heating medium temperatures within ten minutes. The diameter of the pasta was very small, 0.0056 cm.

The use of volumetric heating, such as microwave heating, along with surface heating, such as convective air, is an alternative to surface heating alone to obtain isothermal conditions within a relatively large sample. Temperature profiles of bread samples during microwave heating show that isothermal conditions were obtained in less than one minute and maintained throughout drying, as shown in Figure 2.[76] The isothermal apparatus consisted of a microwave oven with continuous variable power, temperature and air velocity controlled convective hot air, feedback temperature controller linked to the power generator, an analytical balance to monitor weight loss, and a polycarbonate drying tube where the convective hot air flow through and the sample suspended into from underneath the analytical balance. The temperature within the sample was measured using a fiber optic temperature sensor, and the center temperature was shown to be the best location for a cylindrically-shaped sample to control the microwave power to obtain and maintain the desired sample temperature. The computer communicated with a mechanical relay to turn on and off the magnetron. When the sample temperature reached the high limit, a relay connecting the power was switched off, and when the temperature dropped to the lower limit, the relay was turned on again. The high and low limits were usually within 0.3°C of the target temperature. The tightness of the temperature control using a feedback temperature controller depended upon the microwave power. Too high of power resulted in temperature spiking above the desired temperature, and too low of power resulted in the temperature not reaching the desired temperature fast enough. The optimal microwave power to be applied relied on the sample size as well as the moisture content of the sample. As the sample dried, the power level was manually reduced. The surface air temperature needed to be approximately 15°C above the desired sample temperature at the beginning to counter evaporative cooling effects. The air temperature was reduced as the evaporative cooling effects diminished until the air temperature was reduced to the desired sample temperature, which was within 5 min into the drying. The microwave power at the start of drying was 1050 W and significantly reduced to 400 W after 5 minutes and further reduced as the convective air flow governed the isothermal temperature control. Once the convective air flow was reduced to the desired sample temperature the time increments that the power remained off increased until completely off for the last half of drying.

Once the proper schedule of when to turn down the air temperature and microwave power during isothermal drying were determined and reproducibility shown, drying experiments were conducted using just one probe at the center of the sample. A probe at the center did not interfere with weight loss measurements.[76] Thus, three probes were used to verify isothermal conditions at the center, half-radius and near surface (Figure 2a) and top center, mid center and bottom center (Figure 2b) with the mid center probe used for the power control, and then one probe at the center was used for the temperature control during weight loss measurements. For a highly porous materials, such as bread, the isothermally measured effective diffusivity did not result in accurate moisture transfer; however, moisture profile analysis during isothermal drying revealed uniform moisture

(a)

(b)
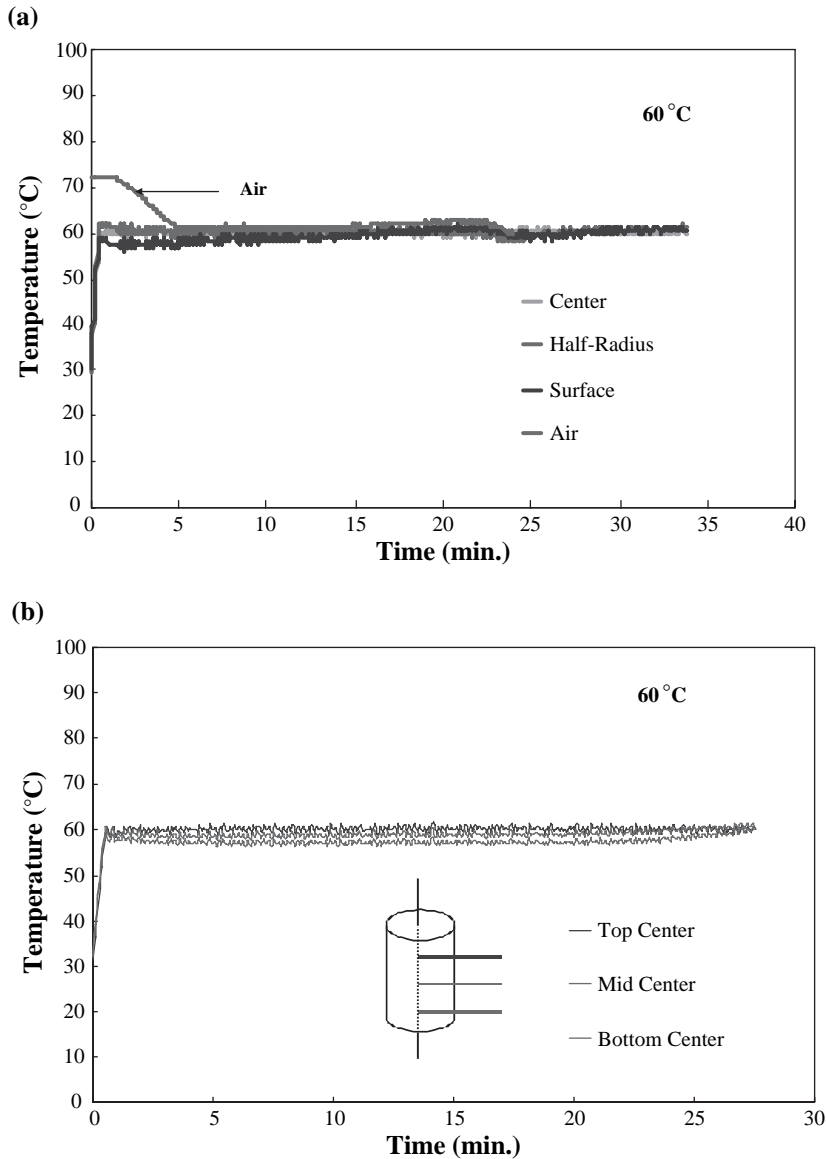
Figure 2 Isothermal temperature profiles of bread at 60°C: a) radial profile; b) longitudinal profile. Adapted from Roberts and Tong.[76]

loss, which differs from a diffusion controlled profile. As stated earlier, the likely mechanism of moisture loss during isothermal drying of highly porous material is evaporation.

Isothermal conditions were also obtained and maintained during drying of potato and carrot, representative hygroscopic low porous materials,[114] as well as for apple, representative hygroscopic developing porous material.[115] The isothermal apparatus used for potato, carrot, and apple was improved by incorporating a PID control for the feedback temperature control scheme. For low porous potato and carrot, the isothermally measured effective diffusivity and corresponding temperature dependence were used to model

convective hot air drying of potato and carrot, and the results showed accurate predictions (<8% error) of moisture profiles during convective hot air drying.[120] What was also striking about these results was that shrinkage and concentration dependence were not taken into account. It was concluded that though accounting for shrinkage and concentration dependence could slightly improve the predictions, the most significant factor in improving the diffusion model prediction is the validation of isothermal conditions during diffusion coefficient measurement. Therefore, establishing isothermal drying conditions can provide both insight into mechanisms of moisture transfer as well as measurement of temperature-dependent parameters.

Effect of Moisture Content on D_{eff} Measurements

Drying rate curves have been shown to deviate from the theoretical prediction in the range of very low moisture contents where the diffusion theory should be valid.[57] Many researchers suggested that diffusivity is not constant, but varies with moisture content.[40,42,59,121] In heat transfer, conduction equations may be used with sufficient accuracy using average values of thermal diffusivities but this is not so in diffusion mass transfer because there is a continuous change in the structure of the material itself as drying proceeds.[42] D_{eff} values during the last stages of the second falling-rate period of drying for various foods were found to be about four to eight times lower than those in the first falling-rate period.[11,27] With moisture dependence, diffusion coefficients were calculated based on an appropriate solution of equation (13) over each segment of the drying period considered.[47,122]

The assumption of a constant, average diffusion coefficient value is valid within the moisture content range where all drying curves at a given temperature merge into one curve (regular regime curve) regardless of initial moisture content.[123,124] Schoeber[124] developed a regular regime method to determine concentration dependent diffusion coefficient for systems in which the moisture diffusivity decreases with decreasing moisture content below the critical moisture content or for cases where the drying rate is governed by mass transfer inside the material. "Regular Regime" was first introduced by Luikov[123] for heat transfer but can be applied to mass transfer. The whole drying process of drying may be divided into three stages. In the first stage, the main role is played by the initial moisture distribution in the following moments. This period is also called the penetration period. The second stage is referred to as the "regular regime". The moisture distribution inside the material does not depend on the initial distribution. The third stage corresponds to steady state in which the moisture content at all points is equal to the ambient moisture content. A requirement for the application of this technique is knowledge of the regular regime curve, which has been determined experimentally at the desired temperature for the case of constant surface concentration. The concentration dependent D_{eff} at one temperature can be calculated from one sorption experiment using the regular regime method. Another advantage of the regular regime method is that this technique can be applied to systems with any degree of shrinkage since a reference component mass centered coordinate is used. The regular regime method has been successfully applied to measure a number of liquid and sold foods with reasonable accuracy.[117,118,125] If the falling rate period of drying falls into this range, a diffusion model developed based on Fick's second law with constant diffusivity can be used to describe the moisture transfer in this drying period.[34,37,102,126] However, during the second falling rate period below moisture content of 0.2 kg/kg dry solids, the diffusivity estimated by the method of slopes of the drying

curves changed substantially with moisture content thus leading to differences between experimental data and model predictions.[104]

The diffusion coefficient's moisture dependence has normally been determined from the experimental drying curves of the average moisture loss with time. As been reported by Crank,[37] the effective diffusion coefficient has been a function of moisture concentration by the following three relationships: linear, $D_{eff} = 1 + aM$; exponential, $D_{eff} = e^{aM}$; and power law, $D_{eff} = M^a$. However at the onset of determining the moisture concentration dependence, the functional relationship is usually not known. Therefore, a series of trial-and error methods is required to determine the function by best-fit analysis.[127] Since average moisture content does not take into account material heterogeneity, the problem of accurately determining the effective diffusivity as a function of moisture concentration using only average moisture content of the material has been well known. A direct method of deriving the diffusion coefficient from experimental moisture profiles in the material during drying is the better approach to accurately determine the moisture dependent effective diffusivity.[82] Magnetic Resonance Imaging (MRI) has been used to obtain transient moisture profiles with high accuracy in complex materials, thus allowing better determination of the effective diffusivity as a function of local moisture contents varying with time and position. In addition to MRI, scanning neutron radiography was also used to obtain moisture profiles of porous materials such as clays from which the effective diffusivity as a function of moisture content can be directly determined.[127] More details of this technique are given in Advanced Methods of Moisture Profile Measurement Section.

Effect of Shrinkage on D_{eff} Measurements

Shrinkage could also have a significant effect on determining the effective diffusivity, as shown by the use of the characteristic dimension parameter in Eq. (39). The thickness of the studied material changes during drying, thus making it is difficult to measure the diffusion coefficient using the Fick's second law solution equation. To solve this problem, Crank[37] and Fish[47] introduced a moving frame of reference in which the thickness remains constant and determined diffusion coefficient of reference. A relationship between the constant-volume (or the proper) diffusion coefficient and the diffusion coefficient of reference was then developed to determine the proper diffusion coefficient. Crank[37] proposed a one-dimensional volume change following the direction of diffusion whereas Fish[47] proposed a three-dimension, isotropic volume change. However, Gekas and Lamberg[128] suggested the concept of arbitrary volume change, which better describes the real volume change. The models for moisture diffusivity developed considering shrinkage are summarized in Table 3.

Shrinkage usually occurs simultaneously with moisture transport in the drying process of foodstuffs and other biological materials.[24] Due to its effect on moisture removal rate and diffusion coefficient as well as other physical properties, such as density and porosity, knowledge of shrinkage phenomenon is necessary to better control the drying process and product characteristics.[23] Negligible shrinkage has usually been assumed to make it easier to solve heat and mass transfer equations but this assumption is valid only for some materials within a limited range of moisture content.[24,129] A model taking the shrinkage effect into consideration will more accurately represent the drying process.[130] Simal et al.[101] found that the D_{eff} values when sample shrinkage was considered and ignored were very different and the drying curve obtained when sample shrinkage was

Table 3 Various models of diffusion coefficients in the case of volume change.

Diffusion coefficient of reference	Diffusion coefficient (proper or constant-volume)	Equation	Reference
D^M	D^V	$\dfrac{D^M}{D^V} = \left(\dfrac{\text{total basic volume}}{\text{actual volume}}\right)^2$	[37]
D'_{pseudo}	D_{proper}	$\dfrac{D'_{pseudo}}{D_{proper}} = \left(\dfrac{\text{dry volume}}{\text{actual volume}}\right)^{\frac{2}{3}}$	[47]
D_{ref}	D	$\dfrac{D_{ref}}{D} = \left(\dfrac{\text{total initial volume}}{\text{actual volume}}\right)^{\frac{2}{n}}$	[128]

taken into account was more accurate. This is also supported by Chen and Johnson[121] who pointed out that negligible shrinkage effect could contribute to the observed discrepancies between experimental data and theoretical values.

The theoretical basis for shrinkage should involve mechanical laws taking into account material stresses and deformations during dehydration. For a complicated food system, this will require knowledge about structure, mechanical and elastic properties of each phase of the system, and variation of moisture content and temperature.[131] Therefore, an experimental approach to investigate food shrinkage is the preferred alternative. Volumetric (bulk) and dimensional shrinkage during drying were determined from changes in volume and dimensions, respectively.[19,24,132]

In vegetables, the volume shrinkage is approximately the same as the volume of water loss during drying in the early stage.[133] It was shown by previous studies that shrinkage of different foods is only dependent on total moisture content under specific operating conditions.[19,133–136] A number of researchers have proposed a simple linear relationship between volumetric or bulk shrinkage (S_b) and moisture content (M) during air drying of vegetables and fruits, such as apple, potatoes, broccoli stems, defined as follows:[22,23,101,131]

$$S_b = aM + b, \tag{44}$$

where S_b is the ratio of the sample volume at any time and the initial volume, V/V_0; and a & b are constants determined at various drying conditions. For some food materials, such as potatoes, apples and garlic, these constant values also depend on the range of moisture content as indicated by two segments of straight lines. For other products, such as carrots and pears, volumetric shrinkage is represented by one linear line in the whole range of moisture contents.[131]

According to Kilpatrick et al.,[134] for the early stages of drying, the bulk shrinkage coefficient (S_b) is related to the moisture content (M) by this following equation:

$$S_b = (M + 0.8)/(M_i + 0.8). \tag{45}$$

Suzuki et al.[133] proposed three alternative models for bulk shrinkage coefficient predictions:

1. *Uniform drying model*. The volume of shrinkage in the sample and the volume of water evaporated are equal. This model results in two equations: one similar to Eq. (44) requiring data for the initial moisture content and bulk density, and the other similar to Eq. (45) requiring data for the equilibrium moisture content and bulk density.
2. *Core drying model*. There exist two parts of the sample: the core (inner part) where the density is equal to the initial value, and the shell (outer part) where the density reaches the equilibrium value for the given drying condition. The proposed equation is:

$$S_b = K\frac{M}{M_i} + 1. \tag{46}$$

3. *Semi core drying model*.- The density of the dried shell is a weighted mean between the initial and equilibrium. The bulk shrinkage coefficient can be predicted from the following equation:

$$S_b = r'\frac{M}{M_i} + n'. \tag{47}$$

Constants K (Eq. 46) and r' and n' (Eq. 47) are determined from empirical equations using initial and equilibrium values of both moisture content and bulk density.[19,133]

In addition to shrinkage, bulk density, particle density, and porosity are important physical properties that are affected by the volume change during drying. Zogzas et al.[20] developed a simple model to predict these properties as a function of material moisture content.

For the model taking shrinkage into account, finite element and finite difference methods have been used to solve the differential equations of mass transfer in a moving boundary problem.[91,92]

Advanced Methods of Moisture Profile Measurement

The traditional approach to analyze moisture transfer mechanism, as described in the previous sections, involves analyzing the mass average moisture loss curves. However, Gekas[62] emphasized the need to analyze moisture profiles to gain greater insight into which mechanism(s) are governing the transfer. A destructive method of determining moisture content that has been used involves immediately freezing the sample in liquid nitrogen at various times during dehydration, allowing the sample to rise towards −4°C in a freezer for cutting, cutting the sample along its characteristic dimension, and then determining the moisture content of each section by the vacuum oven method.[30,81,137,138] Common non-destructive methods for measuring moisture distribution inside a sample are: radiography methods, electric time domain reflectometry (ETDR), fiber optic near-infrared reflectance (NIR), and nuclear magnetic resonance (NMR). The basic concept of radiography is to have a radiation source, such as an accelerator or a reactor, a collimator, which defines the geometric properties of the beam and contains filters, the object sample, and a detector, which records the intensity of the beam after passing through the object.

The source of radiography may be X-rays, gamma rays, or neutrons, where neutron radiography offers the highest hydrogen detection sensitivity.[139] ETDR methods involves a sensing probe which transmits electromagnectic waves and measures the time for the electromagnetic waves to travel through the length of the probe. The velocity of the propagating wave is a function of the material's dielectric properties, which in turn is a function of the material's moisture content.[140] Radiography and ETDR methods are often used to determine moisture distribution in building materials soils, respectively. Fiber optic NIR was developed by Thorvaldsson and Skjöldebrand.[141] Water molecules have absorption peaks in the near infrared region at 0.97, 1.19, 1.45, 1.94, and 2.95 μm, and the basic principle of this measurement is to send light through a fiber optic into the sample and to measure the intensity of the reflected light. This technique shows reasonable results for the moisture distribution in bread samples.[85,141]

NMR is the most common non-destructive method to determine material structure, component saturation, and material properties including self-diffusion coefficient. NMR is based on the measurement of resonant, radio frequency absorption by nonzero nuclear spins in the presence of an applied static magnetic field. Water molecules essentially have four nonzero nuclear spins: proton (^{1}H), deuterium (^{2}H), tritium (^{3}H), and oxygen (^{17}O).[142] Proton has been the most common target nucleus to probe to study moisture transport in foods.[143] Hydrogen atoms of water molecules behave as charged bodies with an angular momentum and thus have a magnetic moment, a vector quantity describing the strength and direction of the magnetic field surrounding the nucleus. An externally applied magnetic field causes these magnetic moments to align with the direction of the external field, which is analogous to dipoles aligning with an externally applied electric field during microwave heating. Nuclei do not align perfectly with the magnetic field and a permanent torque occurs. This torque along with the nuclei's angular momentum results in precession, and the frequency of precession is described by the Larmor relationship:

$$\omega = \gamma \beta_0, \tag{48}$$

where ω is the frequency of precession, or Larmor frequency (radians/s), γ is the magnetogyric ratio, and β_o is the external magnetic field strength (Tesla). During NMR, radio frequency excitation at the Larmor frequency causes magnetic moments to align briefly and then return to equilibrium, which can be recorded by a radio frequency receiver. This return to equilibrium provides information on the physical state of the nuclei, such as mobility and chemical compound identification, and on the local electronic environment of the nuclei, which are all molecule specific. NMR (with imaging), or Magnetic Resonance Imaging (MRI), is a spectroscopic method used to generate internal images based on the magnetic properties of nuclei. Pulsed linear magnetic field gradients are applied to produce a frequency variation across the sample, which can be converted to special coordinates since Larmor frequencies become a function of position. A more detailed explanation of NMR and MRI is provided by Schmidt and Lai.[142]

MRI is capable of providing a measurement of the mass transport by measuring internal variations in component saturations; therefore, it has been used to obtain accurate moisture profiles in model food materials.[108,143] Several researchers used MRI to determine moisture profiles during drying of fruits and vegetables, such as apple, potato, and sweet corn.[144–146] Experimental moisture profiles obtained from MRI measurements can be used to determine the moisture transfer and measuring its corresponding mass transfer

property.[104,108,143] Moisture gradients in fruit have been modeled by Fick's second law and validated by MRI.[147] Frias et al.[148] used MRI to validate diffusion as the mechanism for moisture transfer during drying of rice. MRI has proven to be useful in showing that moisture removal patterns in wheat kernels during drying where a strong function of the grain structural components.[149] Moisture loss analysis showed that the slowest rate of moisture loss occurred in the endosperm, and the greatest moisture removal rate was in the pericarp. The image and gradient vector analysis was also able to provide information on the tendency of moisture movement from endosperm to the embryo through the scutellum epithelium, which functions as an absorption organ, and explains why the embryo retained more moisture after 4 h of drying. Such a study on grains advances our understanding of moisture transfer and will lead to better designed grain dryers. MRI used to study mass transfer in food materials is fast, accurate and nondestructive compared to other methods for determination of the effective diffusivity.[108]

CONCLUSION

The mechanism(s) governing moisture transfer during dehydration is complex and is highly dependent on the food's structure, particularly its porosity, which can change as drying proceeds. Though the use of Fick's second law using effective moisture diffusivity is highly empirical, less so for low porous foods, its use for quantifying moisture transfer will continue until fundamental mechanisms can be determined with certainty and quantified. Therefore, proper measurements of effective moisture diffusivity will be very important in providing as accurate an empirical model as possible. The crucial assumption in determining effective diffusivity experimentally is that isothermal conditions are achieved during drying. One methodology in creating an isothermal drying apparatus was described using controlled volume heating along with controlled surface heating, and the results of the isothermally measured effective moisture diffusivity for low porous foods significantly improved the moisture transfer predictions without taking into account moisture concentration dependence and shrinkage. Finally, in investigating moisture transfer in intermediate and highly porous foods, local equilibrium is often assumed relating water vapor pressure, moisture content, and temperature within a sample. This assumption is certainly an over simplification, and non-equilibrium relationships are needed to be quantified in order to better predict moisture transfer in intermediate and highly porous materials. Methods of measuring moisture and water vapor within foods under drying conditions will continue to be a driving force for advancing our understanding of heat and moisture transfer in solid food materials.

NOMENCLATURE

A   Transfer area, m^2
a_w Water activity
c Concentration, kg/m^3
c_p Specific heat, kJ/kg·K
c_{ds} Specific heat of dry solid, kJ/kg·K
c_v Volumetric heat capacity, kJ/m^3·K
c_w Specific heat of moisture, kJ/kg·K
D_{AB} Binary diffusivity of water vapor in air = 2.6×10^{-5} m^2/s at 25 °C
D_{eff} Effective moisture diffusivity, m^2/s

D_o Diffusivity constant, m^2/s

D' Effective vapor space diffusion coefficient $= \Phi D_{AB}/\tau$, m^2/s

E_a Activation energy, kJ/mol

g Acceleration of gravity, m/s^2

H Relative humidity, $p_w / P^o_w \times 100$

ΔH_d Molar heat of desorption, kJ/kg

h Hydraulic head, m

h_s Surface heat transfer coefficient, W/m^2 K

J_g Liquid flux due to gravity, $kg/m^2 \cdot s$

J_h Heat flux per area, $kJ/m^2 \cdot s$

J_m Mass concentration flux, $(kg/m^3)/ m^2 \cdot s$

J_{liq} Liquid flux, $kg/m^2 \cdot s$

J_{vap} Vapor flux, $kg/m^2 \cdot s$

K Hydraulic conductivity, m/s

k_p Permeability, m^2

k_T Thermal conductivity, $kJ/m \cdot s \cdot K$

L Characteristic dimension along the flow path, m

l Half-thickness from surface in a slab, m

M Moisture content, dry basis, kg H_2O/kg dry solid

M_i Initial moisture content, kg H_2O/kg dry solid

M_∞ Equilibrium moisture content, kg H_2O/kg dry solid

M_t Moisture content at time t, kg H_2O/kg dry solid

N_{Fi} Fick number $= D_{eff} \cdot t / L^2$

n Constant: 0 for planar, 1 for cylinder, 2 for sphere

P Total pressure, kPa

P_D Diffusion coefficient for vapor, $(kg/s) \cdot (m^2) \cdot (kPa/m)$

P^o_w Vapor pressure of water, kPa

p_w Partial pressure of water, kPa

Q Internal energy generation, $kJ/m^3 \cdot s$

R_v Universal gas constant, 82.1 $atm \cdot cm^3/ mol \cdot K$

r Radial dimension from surface in a cylinder or sphere, m

s Slope as defined in equation (38), t^{-1}

T Temperature, K

t Time, second

x Dimension from surface in a slab, m

z Elevation head, m

Greek Symbols

α_T thermal diffusivity, m^2/s

β, γ constants for Eq. (40)

β external magnetic field strength, Tesla

δ thermal gradient coefficient

$\in$ phase conversion factor

Φ porosity or void fraction (fraction of total volume occupied by air or water vapor)

γ magnetogyric ratio

λ latent heat, kJ/kg

μ fluid absolute viscosity, N s/m^2

ρ bulk density of sample, kg $sample/m^3$ sample

ρ_s mass concentration of solid, kg $solid/m^3$ sample

σ, ϖ constants for Eq. (29)

ω frequency of precession, radians/s

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